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MEMORANDUM

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TO: P. L. ROGGENKAMP

FROM: W. E. GRAVES W.E.G.

CALCULATION OF ^{236}Pu CONTAMINANT IN ^{238}Pu PRODUCT

INTRODUCTION

^{238}Pu produced by the irradiation of ^{237}Np contains a small amount of the undesirable contaminant ^{236}Pu. Decay products of the ^{236}Pu give off penetrating γ-radiation. The ^{236}Pu level in normal production is about one ppm. Although this is adequate for some applications, it is too high for others; for all applications a level below one ppm is desirable.

To develop ways to reduce the ^{236}Pu content, mechanisms of its production must be understood quantitatively. Some knowledge of mechanisms has been obtained via test irradiations, but application of this to other situations requires a theoretical framework. This document provides a calculational procedure.

The reactions which lead to ^{236}Pu formation are either ^{237}Np (n,2n) ^{236}Np or ^{237}Np (γ,n) ^{236}Np. Half of the 22-hour half-life ^{236}Np decays to ^{236}Pu. To calculate from first principles the rate of formation of ^{236}Np the following steps are necessary.

- Evaluation of the strengths of sources of particles which lead to the $^{237}Np \longrightarrow {}^{236}Np$ reactions.
- A particle transport calculation to obtain the particle flux in the neptunium due to the specified source.
- Specification, for the materials present, of attenuation cross sections which are appropriate in the above particle transport calculation.
- Determination of the formation rate of ^{236}Np from the particle flux and the (n,2n) and (γ,n) cross sections of ^{237}Np.

Of the above 4 steps, the first 3 are reasonably well in hand but the 4th is not because, although the n,2n cross section is fairly well-known, the γ,n cross section is completely unknown. The (γ,n) cross section will be measured in the near future by the Experimental Physics Division.

The purpose of this memorandum is to describe the details of the above procedure so that

- Sources of the most important uncertainties can be identified and
- The calculations can be carried out on lattices of interest when the γ,n cross section becomes available.

Preliminary calculated results using a fictitious γ,n cross section are given for two mixed lattices in order to illustrate the relative importance of various sources of particles.

SUMMARY

The data and calculation method used lead to too high a prediction of ^{236}Np formation from the n,2n reaction compared to plant experience. The error is perhaps a factor of 2. Various factors could contribute to this error but the likeliest major source of error is the assumed shape of the fission spectrum in the very high energy region. This should be the subject of further study.

In the two mixed lattices studied it is concluded that aluminum in the Np-Al assembly itself constitutes a significantly greater source of contamination than aluminum in the surrounding assemblies. Thus, to the extent that the γ,n reaction is important there is an incentive for reducing the amount of aluminum in the irradiation assemblies.

DISCUSSION

The ratio of ^{236}Pu to ^{238}Pu in a Np target is given by

$$\frac{^{236}Pu}{^{238}Pu} = \frac{f\ NV\left(\phi_\gamma \sigma_{\gamma,n}^{Np} + \phi_n \sigma_{n,2n}^{Np}\right)}{g \cdot h \cdot \sum_E \phi \cdot V\ N\ \sigma_c^{Np}} \qquad (1)$$

where f = fraction of ^{236}Np decaying to ^{236}Pu = 0.5

N = average number density of Np in volume V during irradiation

ϕ_γ = irradiation averaged hard gamma flux in Np

γ_n = irradiation averaged fast neutron flux in Np

g = fraction of ^{238}Np that decays to ^{238}Pu

h = fraction of ^{238}Pu produced that remains at the end of the irradiation.

$\sum_E \phi V\ N \sigma_c$ = total n,γ events in ^{237}Np, averaged over irradiation.

All of the information in the denominator is obtained from normal lattice burnup calculations. The quantities in parentheses in the numerator are the parameters unique to the problem being considered. In particular, ϕ_γ and ϕ_n must be defined more precisely, and computed from particle sources that are to the same normalization as quantities in the denominator.

Particle Sources

The threshold energy for both the γ,n and the n,2n reaction in ^{237}Np is 6.79 Mev.

The only source of neutrons this energetic is the fission process itself. The neutron spectrum above the cutoff should be given to a good approximation by the uncollided fission spectrum. For both ^{235}U and ^{239}Pu the ENDF/B cross section evaluators have chosen the Maxwellian form of the fission spectrum.

$$\chi(E) = \frac{2}{T}\sqrt{\frac{E}{\pi T}}\, e^{-\frac{E}{T}} \qquad (2)$$

where T = 1.273 Mev for ^{235}U and 1.41 Mev for ^{239}Pu. 7.0 Mev

was taken as the practical lower limit for n,2n reactions (n,2n reactions between the threshold and 7.0 Mev are negligible). Using equation 2 the fraction of fission neutrons with energies above 7.0 Mev is 1.14%. The corresponding number for ^{239}Pu is 1.85%. The ^{238}Np fission spectrum is not known and its characteristics were assumed to be the same as those for ^{239}Pu.

Sources of energetic photons include the n,γ reaction on various materials present and also the fission process. There is a great deal of information in the literature on energetic photons from n,γ reactions as well as some recent measurements here.(1) The major contributors in SRP reactors are absorption in aluminum and in iron. For standard irradiations, the target assemblies are sufficiently far from the tank walls so that the iron photons will be negligible compared to the aluminum photons. Literature values for aluminum photons are given in references 2 and 3. There is a strong gamma ray of energy 7.72 Mev. Probably the best value for its source strength is 32 photons per 100 captures.(2) This is thought to be accurate to within 10%. Other gamma rays above the threshold are weak and were ignored. Source strength of energetic photons from the fission process can be estimated from standard sources.(4) For typical SRP lattices this contribution is only about 5% of the contribution from aluminum and hence this was ignored. Thus, the only source of energetic photons considered in this analysis was the 7.72 Mev photons from neutron absorption in aluminum.

Particle Transport Calculation Methods

Of the various approaches to solving the particle transport equation, integral transport theory seems by far the most appropriate approach for this problem. With deuturium dominating the scattering properties of the lattice, most collisions suffered by either the neutrons or photons in the energy range of interest lead to their being degraded below the threshold for the γ,n or n,2n reaction in neptunium. With the calculation thus reduced to a dominantly "first flight collision" calculation a number of integral transport approaches can be considered. For example, standard shielding formulas,(5) with the buildup factor set equal to 1, can be used in connection with the appropriate tabulated functions to estimate such things as the relative contribution of the tank wall and aluminum absorptions in the core in contributing to γ,n reactions in a reflector.(6) In a lattice, St. John(7) used line source integral transport kernels (first order Bickley functions) to estimate relative contributions from neighbors. For the mixed lattice problem, however, more accurate methods are now available in the form of computer programs. In

particular, the INCYCE code[8] is very well suited to this problem. In addition, the CLUCØP code[9] should be useful and accurate for certain applications. INCYCE permits the calculation of particle fluxes in mixed lattice "supercells" in one, two, three or four energy groups. Geometry within each of the cells is restricted to annular geometry. CLUCØP is restricted to one energy group and uniform lattices (repeating cells) but a great deal of geometrical detail may be included within each cell calculation (annular geometry, cluster geometry, azimuthal asymmetry, etc.).

It is unnecessary here to consider the codes themselves in any more detail. However, the conceptual equations solved by each must be considered in order to appreciate the significance of some of the cross sections to be supplied in the next section. If (in the case of neutrons) the second generation source neutrons produced by fission (of the order of 1% of the primary neutron source) is ignored, the integral transport equation for both types of particles can be written as follows

$$\phi_i V_i \Sigma_{t_i} = \sum_j (S_j + \Sigma_{s_j} \phi_j) V_j P_{ij} \qquad (3)$$

The left-hand side of equation (3) represents the number of collisions occurring per second in the i^{th} region. On the right-hand side the quantity in parentheses is the particle source in units of particles per cubic centimeter per second. The product of this and the volume is thus the total source in the j^{th} region in particles per second. P_{ij} is defined as the probability that a neutron originating in the j^{th} region with uniform spatial probability and isotropic angular distribution will have its first collision in the i^{th} region. The summation is conceptually over all region in the lattice (in practice approximations are made at cell boundaries in order to keep the sizes of the P_{ij} matrices low enough to be manageable). The majority of the work involved in solving equation (3) is in computing the collision probability matrix elements P_{ij}. This involves a one or two-dimensional numerical integration. Once these are available the solution of the resulting set of linear simultaneous equations for the fluxes is straightforward.

The fact that isotropic sources are assumed in the process of generating the collision probabilities is important in choosing appropriate values of Σ_s to use on the right-hand side of equation 2. The normal correction made, in the case of neutrons, to account for the fact that scattering in the laboratory system is not isotropic for light elements is to replace Σ_s by $\Sigma_s \cdot (1 - \bar{\mu})$ and correspondingly to replace Σ_t by $\Sigma_a + \Sigma_s \cdot (1 - \bar{\mu})$, where $\bar{\mu} = 2/3A$. This use of the "transport approxi-

mation" assumes however that the scattering is isotropic in the center of mass system.

This is incorrect for neutrons in the energy range being considered. Furthermore, the photon scattering process must be considered completely separately. These considerations are discussed in the next section.

CROSS SECTIONS

Neutrons

The single energy group chosen for cross section averaging was from 7 to 12 Mev. Only in the case of deuterium was a careful evaluation performed. For the other isotopes it was felt to be sufficiently accurate to take average cross sections from group 1 of the 54-group epithermal HAMMER cross section library (7.79 to 10 Mev). The absorption cross section is taken to be the normal absorption cross section plus the inelastic scattering cross section. The scattering cross section was taken to be the P_0 component of the elastic scattering cross section minus the P_1 component. The reduction was substantial even for heavy elements because of the predominantly forward scattering in the center of the mass system at these energies. It should be noted however that degradation in energy due to collision with isotopes other than deuterium has been ignored.

In deuterium the only significant collision processes are n,2n events and elastic scattering. The absorption cross section was taken to be the sum of the n,2n cross section and that portion of the elastic cross section which results in the neutron being degraded in energy below the 7 Mev cutoff. In order to compute the latter quantity it is necessary to know the angular distribution of scattering in the center of mass system as a function of energy. Fortunately, two simplifications appear on consulting the ENDF/B cross section data for deuterium.

- The angular distribution in the center of mass system is almost identical over the entire energy range 7 to 12 Mev, and
- Only the first two Legendre components contribute significantly.

To a good approximation the cross section can be written

$$\sigma(E,\mu) = \sigma(E)\left[1/6 + (1/2)\mu + u^2\right]$$

where μ is as usual the cosine of the scattering angle in the center of mass system. The desired cross section giving the portion of the elastic scattering which leads to degradation

below 7 Mev is then given by

$$\sigma = \frac{\int_{E_L}^{E_u} dE\ \chi(E) \int_{-1}^{\mu(E)} d\mu\ \sigma(E,\mu)}{\int_{E_L}^{E_u} dE\ \chi(E)} \qquad (4)$$

where $\chi(E)$ is given by equation (2) and $\mu(E)$ is the solution of the equation

$$\frac{E_L}{E} = \frac{1}{2}\left[(1+\alpha) + (1-\alpha)\,\mu(E)\right] \qquad (5)$$

Using ENDF/B values of $\sigma(E)$ a numerical integration yields $\sigma = 0.71$ barns.

Adding the n,2n cross section to this yields a total "absorption" cross section of 0.82 barns.

The cross section for elastic collisions which result in neutron retaining an energy greater than 7 Mev is about 0.4 barns. The scattering for these events is peaked very strongly forward however, so that only a small fraction of this should be used in an isotropic scattering code. In this study it was set equal to zero.

The resulting set of one group cross sections for materials of most interest are given in Table I.

Photons

At photon energies of 7.72 Mev the photo electric effect may be ignored and Compton events and pair production compete as attenuation mechanisms. A good summary of the cross sections, angular dependence, energy dependence, etc., of these two processes is given in reference 10. Pair production removes the high energy photon from the energy region above the γ,n threshold and hence acts as a pure absorption process. 95% of the Compton events degrade the photon below the threshold energy and thus also act as "absorption". The 5% of the Compton scattered photons which remain above the γ,n threshold

have a maximum scattering angle in the laboratory system of 5 degrees. It should be an excellent approximation therefore to assume that these scattering events have not occurred and to set the scattering cross section equal to 0. From reference 10 the absorption cross section per atom for photons of 7.72 Mev energy is given by

$$\sigma_a = \underset{\text{(Compton)}}{0.95 \times 0.0618Z} + \underset{\text{(Pair)}}{0.00174Z^2} \text{ barns/atom.}$$

The resulting cross sections for common reactor materials are given in Table II.

Setting all of the scattering cross sections equal to 0 simplifies the photon transport problem represented by equation (3). In fact, it has been reduced to the problem of specified fixed sources in purely absorbing media. The contributions from the various sources on the right-hand side of equation (3) could be added up by hand once the collision probabilities are known.

Cross Sections for n,2n and γ,n Reactions in ^{237}Np

The differential cross section for n,2n events in ^{237}Np in the ENDF/B compilation is a calculated one, but it is in fair agreement with the single measured point in BNL-325. Averaging the cross section over a fission spectrum in the energy range 7 to 12 Mev, using the ETØJ code, yields an n,2n cross section value of 0.15 barns.

The γ,n cross section is not at all well-known. For illustrative purposes in the examples to be considered next a value of 0.10 barns was assumed. This is a reasonable guess based on values for nearby elements.

EXAMPLES

Two examples of mixed lattice irradiation of neptunium assemblies were considered in order to illustrate the calculation procedures, indicate relative importance of certain mechanisms and identify areas of greatest inaccuracies. The first example, called lattice I is a Mark 14-30A lattice with one Mark 30A per hex replaced by a Mark 52 neptunium assembly containing 150 g/ft of ^{237}Np. The second example, called lattice II, is a more complicated mixed lattice illustrated in Figure 1. In this lattice the neptunium target assemblies are clumped and surrounded by heavy water in order to reduce the n,2n flux from neighbors. Nominal specific powers in Mark 14 assemblies were assumed and burnup calculations (using HAMBUR) were run

such that the product quality at the end of the irradiation was 90% (90% of the plutonium was ^{238}Pu). An infinitely repeating mixed lattice was assumed for these calculations. Reaction rates were averaged over the irradiation time and these in turn were used in connection with the results given in the first section to obtain cycle averaged source strengths for both fast neutrons and photons. These results are given in Table III in the form of total assembly source strengths. In the calculations these sources were broken up into much more geometrical detail. For example, the photon sources in the Mark 52 assembly were broken into five separate spatial regions; the inner and outer housing and the three neptunium aluminum tubes.

The above sources were used as input to single energy group INCYCE calculations, using the attenuation cross sections of either Table I or Table II. Again infinitely repeating mixed lattices were assumed. The resulting absolute fluxes in the neptunium regions were multiplied by the appropriate number densities and cross sections for the formation of ^{236}Np as indicated in equation (1). The cycle averaged value for reactions leading to ^{238}Pu was obtained from the HAMBUR output to the same normalization as in Table III. The parts per million $^{236}Pu/^{238}Pu$ were computed from the above results, using $g = 0.91$ and $h = 0.90$ in equation (1), and are shown in Table IV.

Plant experience indicates that the total parts per million in lattice I should be 0.9 - 1.0 parts per million. Furthermore, there are semi-quantitative indications from plant experience that in lattice I the γ,n and n,2n reactions should be of roughly equal importance. Thus, all the calculated results in Table IV are too high. In the case of the γ,n events one can only conclude that the assumed 100 millibarn cross section for γ,n events is much too large. The over-calculation of n,2n events, however, is not so easy to explain since all the required pieces of data are known to at least some degree of accuracy. The most likely sources of error in this case seem to be the following:

1. The integral of the fission spectrum above 7 Mev compared to the total integral of the fission spectrum. This quantity would certainly not have been considered of much importance by the evaluators who produced the recommended fission spectra for the ENDF/B compilation. The use of an alternative functional form, as for example in Glasstone and Edlund, would reduce the integral above 7 Mev by about 30%. The experimental data in this energy range is not particularly good and the whole question requires re-evaluation.

2. The oxygen cross sections in Table I are crude and should be re-evaluated. This is not trivial since the cross section is varying rapidly in the energy range of interest.

3. The angular distribution assumptions made at cell boundaries in INCYCE could be causing significant error in lattice I for neutrons where the majority of the contributors are neighbors. It appears off-hand however, that a more correct treatment would raise rather than lower the calculated parts per million. This point could be investigated by a more detailed investigation with the CLUCØP code.

In order to get a better qualitative idea of the relative importance of various contributions, the results of Table IV were scaled down according to the assumptions that in lattice I

- The total ppm ^{236}Pu is 0.9
- Contributions from neutrons and photons are equal.

The same scaling factors were then applied to lattice II. Results are given in Table V. It is seen that the only significant reduction in going from lattice I to lattice II is the smaller n,2n contribution from neighbors, and that this reduction should be roughly 0.2 ppm.

TABLE I

Neutron Attenuation Cross Sections

Isotope	"σ_a", Barns	"σ_s", Barns
D	0.82	0
O	0.33	0.30
Al	0.93	0.28
^{235}U	3.46	1.22
^{238}U	3.13	0.68
^{237}Np	4.71	0.76
Bi	2.50	1.
Fe	1.38	0.25

TABLE II

Photon Attenuation Cross Sections

Element	"σ_a", Barns	"σ_s", Barns
D	0.0604	0
O	0.581	0
Al	1.057	0
U	20.13	0
Np	20.51	0
Bi	16.86	0
Fe	2.71	0

TABLE III

Total sources (per assembly) of particles capable of causing (n,2n) or (γ,n) events in ^{237}Np. (Normalization: 1000 neutrons of any energy produced by Mk-14 assembly)

Lattice I

Assembly	Neutron Source	Photon Source
Mk 14	11.44	8.86
Mk 30A	6.02	4.05
Mk 52	1.13	8.99

Lattice II

Assembly	Neutron Source	Photon Source
Mk 14	11.44	8.86
Mk 30A	5.97	4.05
Mk 52	1.01	8.56

TABLE IV

Calculated ^{236}Pu Content (ppm) in Example Cases

Lattice I

Source Location	Source Type	Neutron	Photon	Total
Mark 14 or 30A		0.59	0.86	1.45
Mark 52		0.46	2.13	2.59
Total		1.05	2.99	4.04

Lattice II

Source Location	Source Type	Neutron	Photon	Total
Mark 14 or 30A		0.10	0.35	0.45
Mark 52		0.48	2.68	3.16
Total		0.58	3.03	3.61

TABLE V

Rescaled ^{236}Pu Content (ppm) in Example Cases

Lattice I

Source Location	Source Type	Neutron	Photon	Total
Mark 14 or 30A		0.25	0.13	0.38
Mark 52		0.20	0.32	0.52
Total	≡	0.45	0.45	0.90

Lattice II

Source Location	Source Type	Neutron	Photon	Total
Mark 14 or 30A		0.04	0.05	0.09
Mark 52		0.21	0.40	0.61
Total		0.25	0.45	0.70

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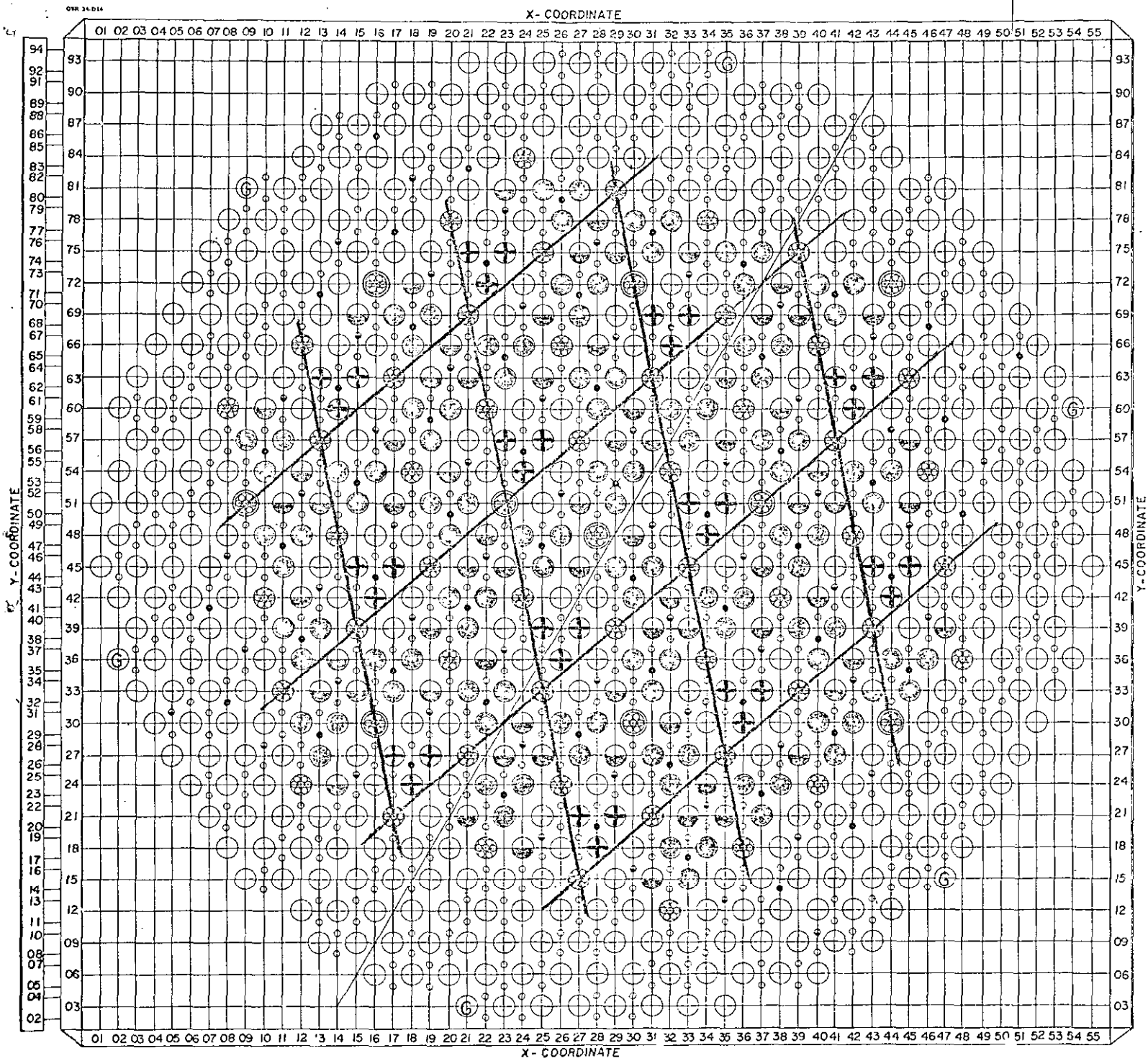

Fig. 1 Lattice II - Lines define supercells which repeat by translation.

Mk 14

Mk 30A

Mk 52

Vacant (in FZ)